

ARPES sensitivity to short-range antiferromagnetic correlations.

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Angle-resolved photoemission spectroscopy (ARPES) is one of most powerful techniques to unravel the electronic properties of layered materials and in the last decades it has lead to a significant progress in the understanding of the band structures of cuprates, pnictides and other materials of current interest. On the other hand, its application to Mott-Hubbard insulating materials where a Fermi surface is absent has been more limited. Here we show that in these latter materials, where electron spins are localized, ARPES may provide significant information on the spin correlations which can be complementary to the one derived from neutron scattering experiments. $\text{Sr}_2\text{Cu}_{1-x}\text{Zn}_x\text{O}_2\text{Cl}_2$, a prototype of diluted spin $S = 1/2$ antiferromagnet (AF) on a square lattice, was chosen as a test case and a direct correspondence between the amplitude of the spectral weight beyond the AF zone boundary derived from ARPES and the spin correlation length ξ estimated from ^{35}Cl NMR established. It was found even for correlation lengths of a few lattice constants a significant spectral weight in the back-bended band is present which depends markedly on ξ . Moreover the temperature dependence of that spectral weight is found to scale with the x dependent spin-stiffness. These findings prove that ARPES technique is very sensitive to short-range correlations and its relevance in the understanding of the electronic correlations in cuprates is discussed.

The discovery of high temperature superconductivity in the cuprates has promoted a renewed interest for the study of electron spin correlations in quantum $S = 1/2$ spin systems. Inelastic neutron scattering (INS) [1] and nuclear spin-lattice relaxation rate ($1/T_1$) [2] measurements have been widely employed to investigate the evolution of spin correlation in several layered materials as cuprates, manganites and pnictides [3] and a series of major breakthroughs obtained. Nevertheless, each one of these techniques has its drawbacks. Inelastic neutrons scattering requires large crystals which are not always available while the determination of the spin correlation length ξ from nuclear magnetic resonance (NMR) $1/T_1$ is not direct but rather based on scaling assumptions. Only when scaling exponents are known, as it is the case for the cuprates, also NMR $1/T_1$ allows a quantitative estimate of ξ . [4]

Remarkably, the onset of sizeable antiferromagnetic (AF) correlations can affect also the band structure. In fact, due to the AF order the Brillouin zone boundary is reduced to half of its size in the ordered state and electron bands bend-back at the AF zone boundary. Hence, it is tempting to use a technique which is quite sensitive to the details of the band features, as it is the case of angle-resolved photoemission spectroscopy (ARPES), to investigate the electron spin correlations. ARPES has allowed to clarify how the Fermi surface of high T_c superconductors (HTSC) evolves with hole doping and which parts are likely involved in the pairing mechanisms, [5] becoming one of the most attractive techniques to study electronic correlations in layered materials. On the other hand, the Charge Density Wave (CDW) instabilities recently detected in HTSC [6–8] are somewhat elusive to ARPES since often they do not give rise to the typical “finger-prints” of CDW order [9, 10]. For example, the

change in the Fermi momentum observed in Bi2201 [11] which could result from a CDW phase is not observed in Bi2212 [12]. Accordingly it has been argued that the absence of a signature of CDW order in ARPES spectra could be likely due to the short range or even dynamic character of the CDWs. Here we show that, on the contrary, ARPES is sensitive to short range electron spin correlations and that it may represent a sensitive probe, complementary to INS, particularly when the correlation length approaches a few lattice steps.

We have chosen $\text{Sr}_2\text{CuO}_2\text{Cl}_2$ (SCOC), one of the best experimental realization of a spin $S = 1/2$ two-dimensional (2D) antiferromagnet on a square lattice, as a test case for ARPES sensitivity to short range spin correlations. This material is characterized by a well defined order parameter and magnetic wave-vector \vec{q} . Moreover, its in-plane AF correlation length, ξ , above the Néel temperature T_N was studied using INS [13] and ^{35}Cl NMR [14]. In addition due to the presence of a natural cleaving plane SCOC is perfectly suited for ARPES experiments.

Many ARPES studies have been carried out on SCOC [15–20] and on the nearly identical $\text{Ca}_2\text{CuO}_2\text{Cl}_2$ system [21, 22], both below and above T_N . As expected, owing to the AF ordering the Brillouin zone boundary is reduced to half of its size in the ordered state and the band bends-back at the antiferromagnetic zone boundary (see inset in Fig. 1). Remarkably, even if this back-bending is believed to be a result of the AF order it persists up to temperatures well above $T_N = 256$ K. This can be explained by the very long AF correlations which are about 100 lattice constants at 300 K [13]. In this work we were able to reduce ξ down to a few lattice constants a , measure with ARPES the intensity in the back-bended part and relate it to a change in the correlation length estimated from ^{35}Cl NMR $1/T_1$ measurements. As it will be

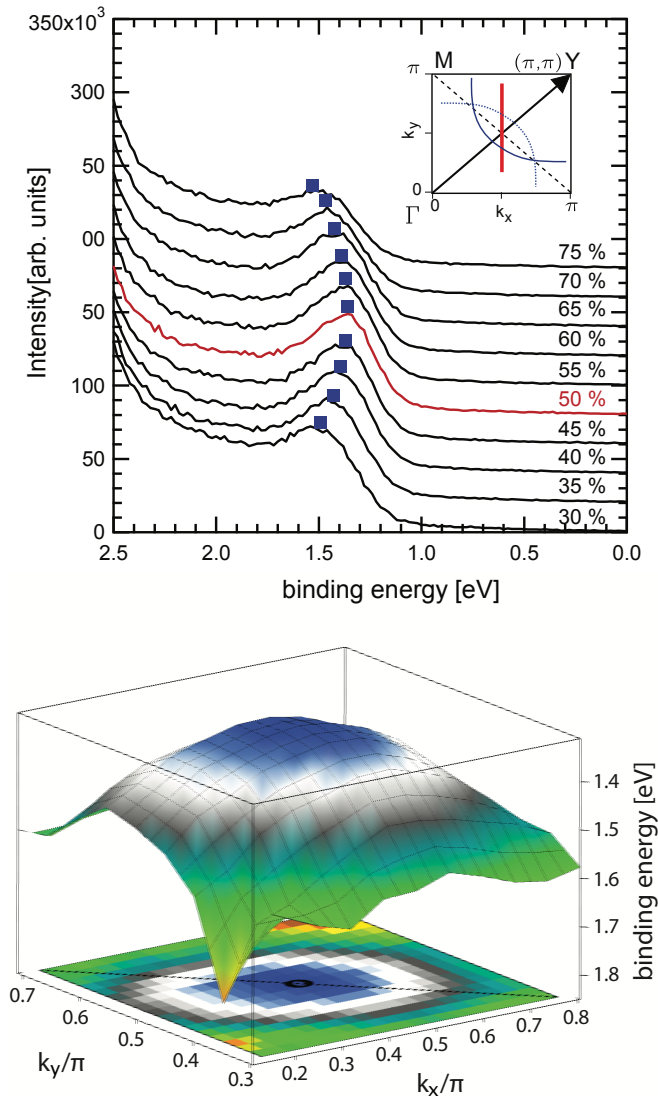


FIG. 1. Upper panel: Band dispersion along the Γ - M direction around the $(\pi/2, \pi/2)$ -point. The position of the peak in each EDC is indicated by blue dots and we find the lowest binding energy at the AFM zone boundary (red line). Inset: A typical cuprate Fermi surface and the folded band upon antiferromagnetic ordering with wave-vector $\vec{q} = (\pi, \pi)$ (black arrow). Lower panel: Extracted dispersion from peak position of several scans in the Γ - M direction. The top of the band moves from the zone corner to $(\pi/2, \pi/2)$ as indicated by the black circle in the 2D projection.

shown in the following some intensity in the back-bended part persists even for $\xi/a \simeq 3$. This is the first systematic study of the sensitivity of ARPES to the AF correlation length which can be relevant also for the understanding of other fluctuating orders.

SCOC single crystals were grown from the melt according to the procedure described by Miller et al. [23]. All ARPES measurements were done with 21.2 eV photons from a monochromized He lamp and a Scienta R4000

electron analyser. Due to the broad band under investigation and for the sake of better statistics, spectra were typically recorded with 20 meV energy resolution and 0.2 degree angular resolution. Fig. 1 shows the dispersion of the highest occupied band at 300 K in the direction indicated by the red line in the inset. A minimal binding energy of about 1.4 eV is found exactly at the AF zone boundary, indicated by the red curve. Beyond that point the band bends-back toward higher binding energies, as expected in view of the new periodicity of the system. At high binding energies around the zone centre we find a waterfall-like feature, typical of many cuprates, whose origin is still under debate [24–26]. We note that since SCOC is insulating, the chemical potential is set by impurity levels and can vary significantly between different samples. For that reason the gap size cannot be measured.

In agreement with previous works we find no sharp peaks even at the lowest binding energy. The origin of the line broadening still needs to be clarified since numerical calculations based on the t-J-model actually predict sharp quasiparticle-like excitations.[17, 27–31] By measuring several momentum cuts and extracting the peak positions for each Energy Distribution Curve (EDC), we were able to map the dispersion of the band over a large area of the Brillouin zone (see Fig 1 lower panel). The dispersion is symmetric around the new zone boundary and has a maximum at $(\pi/2, \pi/2)$. We repeated the measurements at various temperatures between 77 K, deep in the AF ordered phase and up to 450K, nearly twice T_N . We find almost no change in the spectra and, in particular, at all temperatures we observe the back-bended band beyond $(\pi/2, \pi/2)$. However, it should be noticed that in SCOC even at 450K $k_B T \ll J$, the in-plane superexchange coupling, and the correlation length is still more than 20 lattice constants [13].

In order to reduce the correlation length we partially substituted $S = 1/2\text{-Cu}^{2+}$ -ions by spinless Zn^{2+} -ions. In fact, it is well established that in $\text{La}_2\text{Cu}_{1-x}(\text{Zn,Mg})_x\text{O}_4$ T_N can be reduced by diluting the spin lattice [4, 32, 33] and long-range order can even be completely suppressed for substitution levels beyond the percolation threshold (41% for a square lattice). More important for our needs, Zn can significantly reduce the AF correlation length ξ above T_N . We found that Zn can substitute Cu in SCOC crystals up to the substitution limit of 29%, a value below the percolation threshold but large enough to yield a significant reduction of the AF correlations and of T_N . In particular, at this substitution level T_N is reduced to 180 K.

The ARPES spectra of the the spin diluted samples, at all temperatures and at all Zn concentrations, still show intensity beyond the AF zone boundary. In particular, in Fig. 2 we show two normalized ARPES images for the $x=0.29$ sample, each spectrum is normalized using the intensity at high-binding energy around the Γ point and

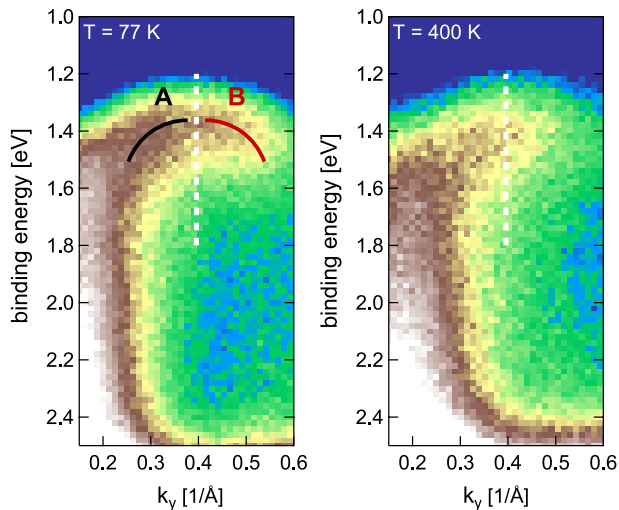


FIG. 2. Comparison of the ARPES images for the $x=0.29$ substituted sample at 77 K (left) and at 400 K (right). The loss of spectral weight beyond the $(\pi/2, \pi/2)$ -point, indicated by the white line, can be observed at high temperature. Nevertheless, the back-bended band is clearly visible, although the correlation length at this temperature is 2-3 lattice constants. The black (A) and red (B) lines represent the band dispersion on both sides of the new zone-boundary at $(\pi/2, \pi/2)$

a constant background, measured at low binding-energy in the Mott gap, was subtracted. In the left panel of Fig. 2 we show an image measured at $T=77$ K in the AF state and on the right the corresponding image at $T=400$ K, where the correlation length is only 3 lattice constants based on the high temperature extrapolation of the NMR data. One can clearly see that, even if at 400 K the intensity of the back-bended part is much weaker than at 77 K, there is still some intensity beyond the AF zone boundary.

In order to provide a quantitative estimate of that intensity we performed the following analysis. We integrate the intensity along the dispersion maximum on both sides of the $(\pi/2, \pi/2)$ -point (dashed white line in Fig. 2) as indicated by the black (A) and red (B) lines. The range of the integration area is 300 meV along the energy axis. Along the momentum axis we choose a range that starts at a point near the Γ -point, where the dispersion is not affected by the waterfall-like feature, and ends at the $(\pi/2, \pi/2)$ -point. An identical range was chosen on the other side of the $(\pi/2, \pi/2)$ -point. Thus, the ratio of the integrals on both sides, B/A , provides a measure of the loss of spectral weight beyond the AF zone boundary.

In Fig. 3(a) we show the ratio B/A as a function of temperature for the pristine and for the $x=0.29$ samples. For the pristine sample the ratio B/A decreases rather slowly with temperature, which is not surprising if one considers that this 2D AF is characterized by a $J \simeq 1450$ K. On the other hand, in the Zn substi-

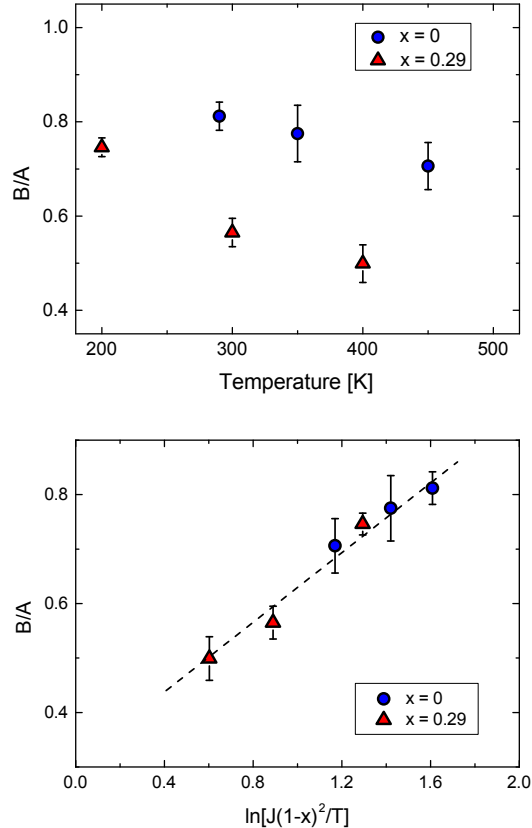


FIG. 3. (a) The ratio of the integrated intensity along the black (A) and red (B) lines in figure 2 as function of the temperature. (b) The ratio B/A is reported as a function of $\ln[J(1-x)^2/T]$ in order to evidence the scaling of its T -dependence with the spin-stiffness.

tuted sample this ratio decreases faster with temperature. In a spin diluted $S = 1/2$ Heisenberg AF on a square-lattice the T -dependence of the in-plane correlation length $\xi \propto \exp(2\pi\rho_s(x)/T)$ is determined by the spin-stiffness $\rho_s(x) = 1.15J(1-x)^2/2\pi$ [4, 32], where the factor $(1-x)^2$ accounts for the probability to find two neighbouring electron spins coupled via the exchange coupling J . Accordingly, one should expect that if B/A ratio is a measure of the spin correlations it should scale as $J(1-x)^2/T$. Remarkably in Fig. 3(b) one observes that such a scaling does occur and that $B/A \propto \ln[J(1-x)^2/T]$. The understanding of this logarithmic functional form goes beyond the aim of the present work.

In order to further evidence the direct relationship between B/A ratio and the in-plane correlation length we quantitatively estimated ξ for different spin diluted samples. ξ can be conveniently derived from nuclear spin-lattice relaxation rate $1/T_1$, an approach successfully used in the analogous $\text{La}_2\text{Cu}_{1-x}\text{Zn}_x\text{O}_4$ compound [4] where a quantitative agreement with the ξ directly

measured by INS was found. In fact, by resorting to scaling arguments, once the hyperfine coupling A and dynamical scaling exponent z are known one can establish a one-to-one correspondence between $1/T_1$ and ξ . [4] In particular, if one considers a 2D $S = 1/2$ antiferromagnet on a square lattice and a nucleus which is essentially coupled just with its nearest neighbour $S = 1/2$ spin, one can write

$$\frac{1}{T_1} = \frac{\gamma^2 A^2 \hbar \sqrt{\pi}}{4 J k_B} \xi^{z+2} \beta(\xi) \int_{BZ} \frac{d\vec{q}}{(1 + q^2 \xi^2)^2}, \quad (1)$$

with γ the nuclear gyromagnetic ratio and $1/\beta(\xi) = \xi^2 \int_{BZ} d\vec{q}/(1 + q^2 \xi^2)$ a factor which preserves the spin sum rule. [4] For $\xi \gg 1$ this expression yields to $1/T_1 \propto \xi^z$. In SCOC one can assume the dynamical scaling exponent $z = 1$ derived from INS [13] and NQR experiments [4] for other cuprates on a square lattice, as La_2CuO_4 . Since A was not very precisely estimated from previous shift measurements, [14] we decided to determine it by taking for A the value which allows to match the absolute value for ξ determined from $^{35}\text{Cl}1/T_1$ and from INS over the same T range. Such a match occurs for $A \simeq 2.7$ kOe, a value very close to the one that can be derived from the high- T limit of $1/T_1$. In the case of diluted samples one has to consider a correction of the average hyperfine coupling by a factor $(1 - x)$ [4], accounting for the probability that ^{35}Cl nuclear spin is coupled to the nearest neighbour Cu^{2+} spin.

The temperature dependence of $^{35}\text{Cl}1/T_1$, determined by using standard radiofrequency pulse sequences, is shown in Fig. 4 for three representative samples. One observes that $1/T_1$ progressively increases upon cooling, shows a peak at T_N and it decreases upon further cooling down. The progressive growth of $1/T_1$ upon approaching T_N from above is associated with the progressive growth of the in-plane AF correlations (see Eq.1). [14] One can now use Eq. 1 to derive $\xi(T, x)$ from $1/T_1$ data. The temperature dependence of $\xi(x, T)$ is shown in Fig. 5. One notices that for the diluted samples the correlation length grows exponential on decreasing temperature, while for the undoped SCOC a crossover to an XY behaviour, characterized by a more rapid increase of ξ , is observed on approaching T_N as pointed out by previous NMR experiments. [14]

In Fig. 6 we show the dependence of ARPES intensity B/A ratio on the correlation length determined from $^{35}\text{Cl}1/T_1$. We find that for both samples the intensity beyond the AF zone boundary as measured by ARPES is controlled by the AF correlation length. For $\xi \leq 15$ lattice units the B/A ratio shows a significant dependence on ξ , which clearly demonstrates that ARPES spectra are indeed sensitive to AF correlations even when they are as short as a few lattice constants. This is an evidence that ARPES besides being a well established technique to investigate the electronic structure of layered materials it allows also to detect short-range electron spin cor-

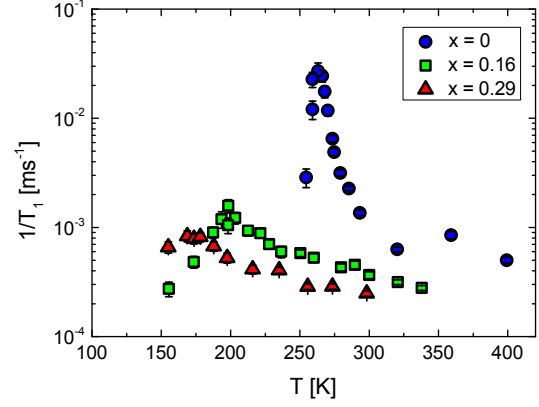


FIG. 4. Temperature dependence of $^{35}\text{Cl}1/T_1$ in $\text{Sr}_2\text{Cu}_{1-x}\text{Zn}_x\text{O}_2\text{Cl}_2$ for $x = 0, 0.16, 0.29$ measured in a 5.6 Tesla magnetic field. T_N is reduced by the Zn substitution as can be measured by following the peak position in $1/T_1$ as the Zn concentration is increased.

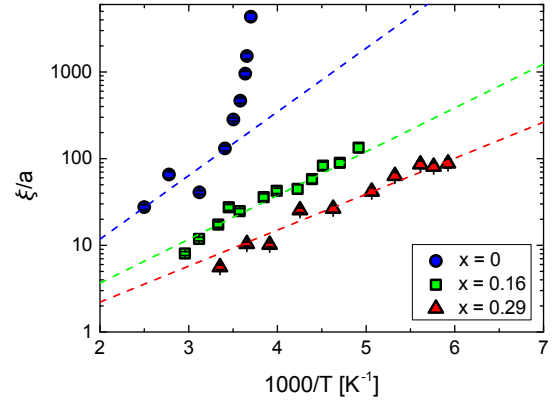


FIG. 5. The AF correlation length in units of lattice constants as function of $1/T$ for three different Zn concentrations. ξ was calculated from the $1/T_1$ data using Eq. 1. The dashed lines show the exponential growth expected for a two-dimensional $S = 1/2$ antiferromagnet. The deviation from that trend observed for $x = 0$ at $T \rightarrow T_N$ was ascribed to a small planar spin anisotropy [14].

relations. For correlation lengths larger than 15 lattice constants we observe almost no change in the spectra and hence it is not possible to distinguish the AF long range order from a fluctuating phase characterized by ξ of the order of tens of lattice constants.

In conclusion, we have shown for the first time that the back-bended part of the highest occupied band changes upon decreasing correlation length. This can be seen as a proof that AF correlations are indeed responsible for this back-bending and that ARPES is a sensitive probe to very short-range AF correlations. Using Zn substitution and by suitably increasing the temperature we were

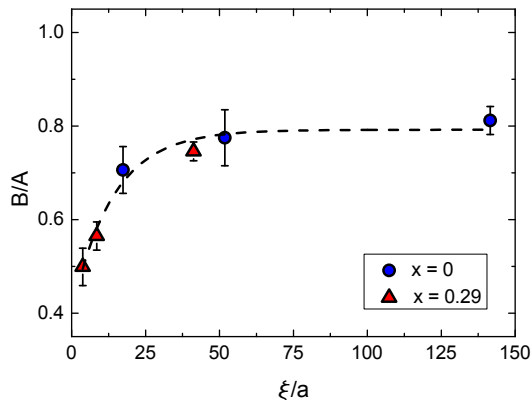


FIG. 6. The ratio B/A is reported as a function of correlation length as measured by NMR data (see figure 5). Dashed line is a guide to the eye.

able to control the correlation length down to 3 constants verified by NMR measurements. Even for the shortest correlation length the spectra surprisingly showed significant spectral weight beyond the AF zone boundary.

The ARPES ability to detect short-range correlations has relevant implications also for other kind of orders. In particular, our findings suggest that the absence of the expected fingerprints of a CDW instability in the ARPES spectra of the under doped cuprates are not simply a consequence of the short range of the CDW correlations.

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- [1] B. Keimer, N. Belk, R. J. Birgeneau, A. Cassanho, C. Y. Chen, M. Greven, M. A. Kastner, A. Aharony, Y. Endoh, R. W. Erwin, and G. Shirane, *Phys. Rev. B* **46**, 14034 (1992).
- [2] A. Rigamonti, F. Borsa, and P. Carretta, *Reports on Progress in Physics* **61**, 1367 (1998).
- [3] G. S. Tucker, D. K. Pratt, M. G. Kim, S. Ran, A. Thaler, G. E. Granroth, K. Marty, W. Tian, J. L. Zarestky, M. D. Lumsden, S. L. Bud'ko, P. C. Canfield, A. Kreyssig, A. I. Goldman, and R. J. McQueeney, *Phys. Rev. B* **86**, 020503 (2012).
- [4] P. Carretta, A. Rigamonti, and R. Sala, *Physical Review B* **55**, 3734 (1997).
- [5] A. Damascelli, *Review of Modern Physics* **75**, 473 (2003).
- [6] T. Wu, H. Mayaffre, S. Kramer, M. Horvatic, C. Berthier, W. N. Hardy, R. Liang, D. A. Bonn, and M.-H. Julien, *Nature* **477**, 191 (2011).
- [7] G. Ghiringhelli, M. Le Tacon, M. Minola, S. Blanco-Canosa, C. Mazzoli, N. B. Brookes, G. M. De Luca, A. Frano, D. G. Hawthorn, F. He, T. Loew, M. M. Sala, D. C. Peets, M. Salluzzo, E. Schierle, R. Sutarto, G. A. Sawatzky, E. Weschke, B. Keimer, and L. Braicovich, *Science* **337**, 821 (2012), <http://www.sciencemag.org/content/337/6096/821.full.pdf>.
- [8] J. Chang, E. Blackburn, A. T. Holmes, N. B. Christensen, J. Larsen, J. Mesot, R. Liang, D. A. Bonn, W. N. Hardy, A. Watenphul, M. v. Zimmermann, E. M. Forgan, and S. M. Hayden, *Nat Phys* **8**, 871 (2012).
- [9] V. Brouet, W. L. Yang, X. J. Zhou, Z. Hussain, N. Ru, K. Y. Shin, I. R. Fisher, and Z. X. Shen, *Phys. Rev. Lett.* **93**, 126405 (2004).
- [10] E. Lahoud, O. N. Meetei, K. B. Chaska, A. Kanigel, and N. Trivedi, *Phys. Rev. Lett.* **112**, 206402 (2014).
- [11] M. Hashimoto, *Nature Phys.* **6**, 414 (2010).
- [12] A. Kanigel, U. Chatterjee, M. Randeria, M. R. Norman, G. Koren, K. Kadowaki, and J. C. Campuzano, *Phys. Rev. Lett.* **101**, 137002 (2008).
- [13] M. Greven, R. J. Birgeneau, Y. Endoh, M. A. Kastner, B. Keimer, M. Matsuda, G. Shirane, and T. R. Thurston, *Physical Review Letters* **72**, 1096 (1994).
- [14] F. Borsa, M. Corti, T. Goto, A. Rigamonti, D. C. Johnston, and F. C. Chou, *Physical Review B* **45**, 5756 (1992).
- [15] B. O. Wells, Z. X. Shen, A. Matsuura, D. M. King, M. A. Kastner, M. Greven, and R. J. Birgeneau, *Physical Review Letters* **74**, 964 (1995).
- [16] S. LaRosa, I. Vobornik, F. Zwick, H. Berger, M. Grioni, G. Margaritondo, R. J. Kelley, M. Onellion, and A. Chubukov, *Physical Review B* **56**, R525 (1997).
- [17] J. J. M. Pothuisen, R. Eder, N. T. Hien, M. Matoba, A. A. Menovsky, and G. A. Sawatzky, *Physical Review Letters* **78**, 717 (1997).
- [18] C. Dürr, S. Legner, R. Hayn, S. V. Borisenko, Z. Hu, A. Theresiak, M. Knupfer, M. S. Golden, J. Fink, F. Ronning, Z.-X. Shen, H. Eisaki, S. Uchida, C. Janowitz, R. Müller, R. L. Johnson, K. Rossnagel, L. Kipp, and G. Reichardt, *Physical Review B* **63**, 14505 (2000).
- [19] S. Haffner, C. G. Olson, L. L. Miller, and D. W. Lynch, *Physical Review B* **61**, 14378 (2000).
- [20] S. Haffner, D. M. Brammeier, C. G. Olson, L. L. Miller, and D. W. Lynch, *Physical Review B* **63**, 212501 (2001).
- [21] F. Ronning, C. Kim, D. L. Feng, D. S. Marshall, A. G. Loeser, L. L. Miller, J. N. Eckstein, I. Bozovic, and Z.-X. Shen, *Science* **282**, 2067 (1998).
- [22] F. Ronning, K. Shen, N. Armitage, a. Damascelli, D. Lu, Z.-X. Shen, L. Miller, and C. Kim, *Physical Review B* **71**, 094518 (2005).
- [23] L. L. Miller, X. L. Wang, S. X. Wang, C. Stassis, D. C. Johnston, J. Faber, and C.-K. Loong, *Physical Review B* **41**, 1921 (1990).
- [24] D. S. Inosov, J. Fink, A. A. Kordyuk, S. V. Borisenko, V. B. Zabolotnyy, R. Schuster, M. Knupfer, B. Büchner, R. Follath, H. A. Dürr, W. Eberhardt, V. Hinkov, B. Keimer, and H. Berger, *Physical Review Letters* **99**, 237002 (2007).
- [25] S. Basak, T. Das, H. Lin, J. Nieminen, M. Lindroos, R. S. Markiewicz, and A. Bansil, *Physical Review B*, 214520.
- [26] J. Graf, G.-H. Gweon, K. McElroy, S. Y. Zhou, C. Jozwiak, E. Rotenberg, A. Bill, T. Sasagawa, H. Eisaki, S. Uchida, H. Takagi, D.-H. Lee, and A. Lanzara, *Physical Review Letters* **98**, 67004 (2007).
- [27] C. Kim, F. Ronning, A. Damascelli, D. L. Feng, Z.-X. Shen, B. O. Wells, Y. J. Kim, R. J. Birgeneau, M. A. Kastner, L. L. Miller, H. Eisaki, and S. Uchida, *Phys. Rev. B* **65**, 174516 (2002).
- [28] R. B. Laughlin, *Physical Review Letters* **79**, 1726 (1997).
- [29] K. M. Shen, F. Ronning, D. H. Lu, W. S. Lee, N. J. C.

- Ingle, W. Meevasana, F. Baumberger, A. Damascelli, N. P. Armitage, L. L. Miller, Y. Kohsaka, M. Azuma, M. Takano, H. Takagi, and Z.-X. Shen, Physical Review Letters **93**, 267002 (2004).
- [30] G. Sangiovanni, O. Gunnarsson, E. Koch, C. Castellani, and M. Capone, Physical Review Letters **97**, 46404 (2006).
- [31] A. S. Mishchenko and N. Nagaosa, Physical Review Letters **93**, 36402 (2004).
- [32] O. P. Vajk, P. K. Mang, M. Greven, P. M. Gehring, and J. W. Lynn, Science **295**, 1691 (2002).
- [33] P. Carretta, G. Prando, S. Sanna, R. de Renzi, C. Decorse, and P. Berthet, Phys. Rev. B **83**, 180411 (2011).